



# Overstory vegetation influence nitrogen and dissolved organic carbon flux from the atmosphere to the forest floor: Boreal Plain, Canada

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## ABSTRACT

Nitrate, ammonium, total dissolved nitrogen (TDN), dissolved organic nitrogen (DON) and dissolved organic carbon (DOC) concentrations and flux were measured for one year in bulk deposition and throughfall from three stand types (upland deciduous, upland conifer and wetland conifer) on the Boreal Plain, Canada. Annual (November 2006 to October 2007 water year) flux rates in bulk deposition were 80, 216, 114 and 410 mg N m<sup>-2</sup> for nitrate, ammonium, DON and TDN, respectively, and 3.5 g C m<sup>-2</sup> for DOC. The nitrate and ammonium flux in throughfall were approximately 50% of the flux in bulk deposition, while TDN flux in throughfall was 60–74% of the flux in bulk deposition. The DOC flux in throughfall was approximately 2 times greater than DOC flux in bulk deposition, while there was no detectable difference in DON flux. The forest canopy generally had the most impact on throughfall chemistry during the active growing season as compared with the dormant season, although DOC concentrations in throughfall of deciduous stands was highest during autumn. For the upland stands, TDN flow-weighted mean concentrations in the snowpack were not detectably different from the concentrations in throughfall and bulk deposition throughout the rest of the year. However, ammonium concentrations were lower and DON concentrations were higher in the snowpack than in either throughfall or bulk deposition for the other seasons, suggesting some transformation of ammonium to DON within the snowpack.

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## 1. Introduction

Although many temperate forests are naturally nitrogen (N) limited (Shaver and Chapin, 1980; Vitousek and Howarth, 1991), much of the research on N deposition, cycling and export in forested ecosystems (see Gundersen et al., 1998; Michalzik et al., 2001 for summaries) has been conducted in areas of potential N saturation (e.g., northeastern United States and central Europe). Nitrogen saturation describes changes to forest soil N dynamics that occur when the supply of N is so large that bioavailable N is in excess of biotic demand (Aber et al., 1989). It implies that other factors, such as light or other nutrients have become more limiting than N. Knowledge is lacking in terms of baseline N deposition in regions with low N deposition, such as the Boreal Plain. This ecozone in western Canada, covers about 650 000 km<sup>2</sup>, an area greater than Finland and Norway combined (Fig. 1). Although there are some estimates of N flux in bulk deposition within this vast region (Alberta Environment, 2008; Kochy and Wilson, 2001; Shaw

et al., 1989), none of these previous studies measured the effect of the forest canopy on N flux to the forest soils. As a result, knowledge of how forest canopy types affect N flux in throughfall for this region is lacking.

Much of the Boreal Plain is quite remote and, historically, has not received high inputs of N from industrial or agricultural sources. However, oil production from oil sands in the province of Alberta increased more than 4-fold during the last 10 years, and is expected to increase another 10-fold by 2015 (Severson-Baker et al., 2008). Increased production, combined with changes in mining procedures, has led to a modeled 5-fold increase in nitrogen oxides (NO<sub>x</sub>) emissions from approximately 60 Mg day<sup>-1</sup> in 1990 to a projected rate of 300 Mg day<sup>-1</sup> by 2012 (Golder Associates, 2002). Much of the emissions is related to the heavy machinery used for extraction of the oil sands, however many of the mines also upgrade the bitumen on site, which results in further NO<sub>x</sub> emissions. There was also a 7-fold increase in coalbed methane extraction from 2003 to 2006 within the province of Alberta (Government of Alberta, 2009). The effects of the cumulative industrial development will likely impact the inorganic N (IN = nitrate [NO<sub>3</sub><sup>-</sup>] + ammonium [NH<sub>4</sub><sup>+</sup>]) and total N deposition on the Boreal Plain, making it essential to examine current rates of N deposition.

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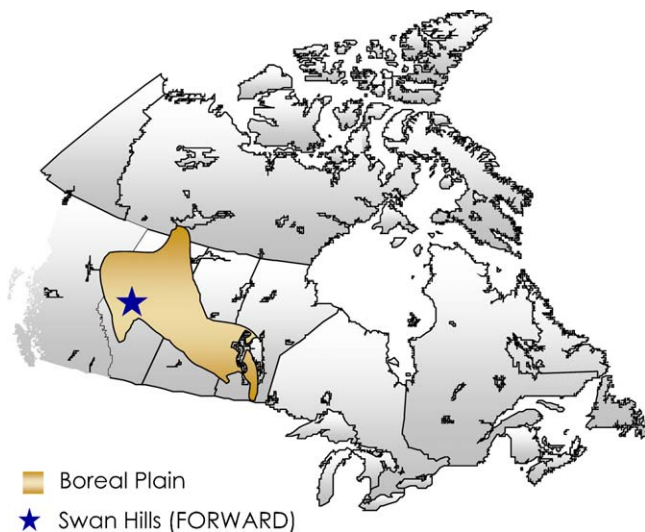


Fig. 1. Location of the Swan Hills, Alberta, Canada.

Throughfall (precipitation passing through the forest canopy) and stemflow (precipitation dripping down plant branches and stems) chemistry and flux tend to be related to the flux in incoming bulk deposition. In areas with greater annual N deposition rates, throughfall and stemflow may be enriched with N (Dise et al., 1998). In areas of relatively low N deposition however, forest canopies generally retained N (Duchesne and Houle, 2006; Friedland et al., 1991; Piirainen et al., 1998; Potter et al., 1991). In fact, negligible throughfall N fluxes generally occur when annual IN deposition rates are less than  $400 \text{ mg N m}^{-2}$  (Dise et al., 1998). Throughfall flux was also correlated with the dominant vegetation type. Morphological differences between conifer and deciduous species (e.g., crown form, leaf shape and cuticle thickness) often result in differences in throughfall and stemflow chemistry and flux by stand type (De Schrijver et al., 2007; Henderson et al., 1977; Michalzik et al., 2001; Verry and Timmons, 1977).

Dissolved organic carbon (DOC) is often correlated to N fluxes in boreal forest soils. For example,  $\text{NO}_3^-$  removal rates in riparian soils in southern Ontario (Devito et al., 2000) and boreal wetland soils in southern Sweden (Davidsson and Stahl, 2000) were positively related with DOC content. Also, C:N ratios in boreal forest floor and soils were positively correlated with N immobilization rates (Cameron and Haynes, 1986; McNulty et al., 1991). As a result, the C:N ratio has been used as an indicator of potential mineralization and nitrification rates, and to predict N losses through IN leaching (McNulty et al., 1991). Therefore, understanding how various soil and stand types affect DOC flux to the soil is important for understanding how different soil and stand types affect N cycling and exports.

This study is a component of the Forest Watershed and Riparian Disturbance (FORWARD) Project, a long-term, multidisciplinary study designed to develop hydrological and water quality models for direct application to industrial forest planning in Boreal Plain watersheds (Prepas et al., 2008a). Studies measuring annual N flux in bulk deposition and throughfall often assist with predicting N export in streams draining watersheds (Dise et al., 1998). In addition, accurate measurements of the differences in N flux above and below the forest canopy in common soil/stand types indicate the forest canopy's role in retaining, transforming or enriching the nutrient flux to the forest soils. The forest canopy can then be entered into these export models as a specific compartment that will assist the FORWARD project with the development of predictive water quality models both in reference and disturbed watersheds. Therefore, the purpose of this study was to: (1)

measure the effect of different forest soil and overstory vegetation types (upland conifer, upland deciduous and wetland conifer stands) on aqueous N and DOC flux to soils in a small ( $15.6 \text{ km}^2$ ) relatively undisturbed watershed on the Boreal Plain; (2) examine the relative amounts of IN and dissolved organic N (DON) comprising the total dissolved N (TDN) flux; and (3) relate the flux rates to patterns of N deposition across North America.

## 2. Methods

### 2.1. Site description

The Willow watershed ( $15.6 \text{ km}^2$ ; Fig. 2) is within the FORWARD project study area in the Swan Hills, located 230 km northwest of Edmonton, Alberta (Fig. 1). Dominant soil orders in the Swan Hills are Luvisolic, Organic and Brunisolic, but Gleysolic and Regosolic soil types also occur (Ecological Stratification Working Group, 1996). The forest vegetation was dominated by trembling aspen (*Populus tremuloides* Michx.), balsam poplar (*P. balsamifera* L.), paper birch (*Betula papyrifera* Marsh.), white spruce (*Picea glauca* (Moench) Voss), lodgepole pine (*Pinus contorta* Dougl. ex Loud. var. *latifolia* Engelm) and balsam fir (*Abies balsamea* (L.) Mill) in well-drained sites and black spruce (*Picea mariana* (Mill.) BSP) and tamarack (*Larix laricina* (Du Roi) K. Koch) in poorly drained sites (Ecological Stratification Working Group, 1996).

Aerial photograph interpretation from 1994 demonstrated that the land cover in the Willow watershed was approximately 53% deciduous-dominated (30% or less conifer crown closure), 39% conifer-dominated forest (70% or more conifer crown closure in the forest canopy), 5% mixed-wood forest (21–69% conifer crown closure), and 3% non-forested. The watershed was relatively undisturbed with limited harvesting (35 ha clear cut in 1980), some road building along the west and south boundaries of the watershed and minor amounts (approximately 1% of the watershed area) of oil and gas exploration (cutting of seismic lines) and extraction (building of access roads and well sites).

The climate is sub-humid (Zoltai et al., 1998) and precipitation is temporally and spatially variable (Pelster et al., 2008). Annual precipitation from 1978 to 2008 ranged from 394 to 777 mm, with a mean of 577 mm (Environment Canada, 2008a). Runoff patterns reflect this variability, with instantaneous discharge in the Willow watershed during 2007 ranging by a factor of more than 100-fold, and annual runoff varying 3-fold between 2002 and 2007. Streamwater pH was near neutral to slightly basic, ranging from 7.2 to 8.2 in the Willow stream during 2007. For more detailed descriptions of the Willow watershed, see Burke et al. (2005) and Prepas et al. (2006, 2008b).

### 2.2. Sampling

Four stands from each of three stand types (upland conifer, upland deciduous and wetland conifer – see Table 1) were randomly selected within the Willow watershed (Fig. 2). Crown closure was measured at 25 points along two 60-m transects set up in a cross formation within each stand (Fig. 2). The points were assigned to four (0–25%, 26–50%, 51–75%, and 76–100%) crown closure classes. Three sampling locations were selected from each stand using stratified random sampling to ensure proportional representation of the crown closure classes.

Throughfall samplers were elevated 0.6 m above the ground and consisted of two 15.2 cm inner diameter polyethylene funnels connected by polyvinyl tubing to a 2 L brown, opaque, high-density polyethylene Nalgene bottle. Sample bottles were partially buried (approximately 80% of bottle below-ground) to keep samples relatively cool and out of the sun. Four bulk deposition collectors were also installed in the open within the watershed

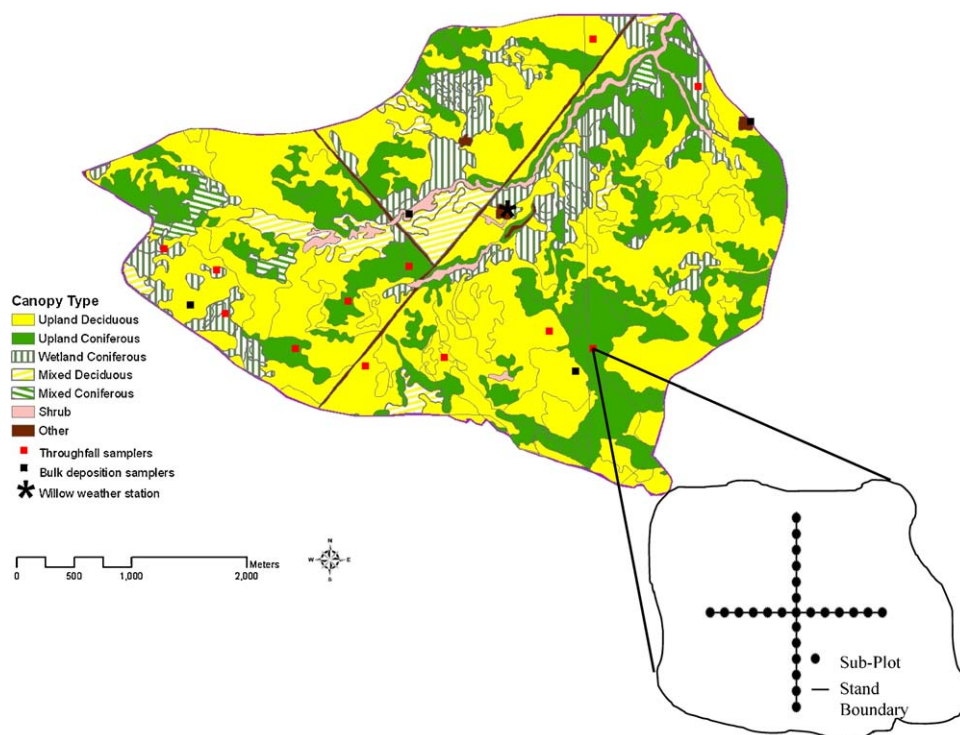


Fig. 2. Forest cover map for the Willow watershed, with throughfall and stemflow sample plot layout.

(Fig. 2): these consisted of a single 25.4 cm inner diameter polyethylene funnel elevated approximately 3 m above the ground and attached to a 2 L brown sample bottle as above. Bulk deposition collectors were situated to ensure minimal effect of forest vegetation (i.e., the slope from the samplers to the top of the adjacent treeline did not exceed 30° from the horizontal). Locations were selected to provide good coverage of bulk open deposition patterns across the watershed area. A Campbell Scientific weather station was installed within the Willow watershed, less than 2 km from the bulk deposition and throughfall sample locations, and provided continuous (every 10 min) monitoring of precipitation volume and air temperature (Fig. 2). Precipitation data from the weather station, along with an Environment Canada weather station located approximately 40 km southeast were used to verify bulk deposition volume measurements and provide additional estimates of precipitation within the watershed.

Throughfall and stemflow collection began in mid-May 2007 and continued throughout the snow-free season (1 May to 31 October). Samples were generally collected every two weeks, although during periods with heavy rains, the sample period was shortened. During the autumn period very little precipitation fell (25.4 mm between 17 August and 31 October), and as a result, only one set of samples could be collected during this time. The volume

of throughfall and bulk deposition was measured and chemistry samples from stemflow, throughfall and bulk deposition were collected approximately every two weeks from May through August. Throughfall and bulk deposition collectors were rinsed with distilled water after each sample was collected. Although the stemflow collectors were persistently damaged by bears, the sample bottles were usually untouched. As a result, even though stemflow volumes could not be determined, the nutrient concentrations could still be accurately measured.

Samples were stored on ice until they could be processed and preserved. Since biocide was not added to the sample bottles and samples were collected every two weeks, bioconversion and immobilization of N could have been a problem, particularly during the warm summer months. However in August, samples were collected on an event basis to test this hypothesis and regression analysis found no relationship ( $n = 80$ ,  $P = 0.88$ ) between the number of days the sample was stored in the sample bottle and percent DON. This suggests that immobilization of IN by microbes in the samples was limited.

The remoteness of the sites and depth of the snowpack made it unfeasible to collect multiple samples during the winter. As a result, similar to the methods used by Kolka et al. (1999), a snowpack survey was completed in early March 2007, when the

Table 1

Site characteristics for dominant stand types in the Willow watershed. Mean species composition of canopy given in brackets.

	Stand type		
	Upland deciduous ( $\leq 20\%$ conifer cover)	Upland conifer ( $\geq 80\%$ conifer cover)	Wetland conifer ( $\geq 80\%$ conifer cover)
Dominant forest vegetation	Trembling aspen (70%) Balsam poplar (10%) Paper birch (10%) Other (10%)	Lodgepole pine (80%) White spruce (10%) Balsam fir (10%)	Black spruce (90%) Lodgepole pine (10%)
Mean % crown closure	85 $\pm$ 10	89 $\pm$ 8	74 $\pm$ 22
Mean age (years)	65	78	65
Mean DBH $\pm$ 1 SD (cm)	18.9 $\pm$ 8.4	24.7 $\pm$ 7.4	11.0 $\pm$ 3.8
Dominant soil orders	Luvisolic; Brunisolic	Luvisolic; Brunisolic	Organic



snowpack is typically at its maximum, as a proxy for throughfall and stemflow to estimate dissolved N flux during the winter season (1 November 2006 to 30 April 2007). This method, while not optimal, was still expected to provide suitable estimates of nutrient flux during winter, since another study found no detectable difference in  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations between snowpack surveys and bulk deposition samplers from the National Atmospheric Deposition Program (Hidy, 2003). Also, while N concentrations in the snow can change while the snow is held within the forest canopy, few alterations occur to snow chemistry below the canopy (Pomeroy et al., 1999). As a result the snowpack survey should be representative of throughfall and stemflow chemistry in this region. The snow water equivalent (SWE) was compared with continuous measurements for snowfall from a Campbell Scientific weather station fitted with a snowfall conversion adapter and located within the Willow watershed to assure that N was not lost due to entrainment in meltwater.

Three snow cores were collected using a copper coring tube (ID = 5.08 cm) at each of nine throughfall/stemflow sample locations (three samples from each stand type) and at three bulk deposition sample locations. Since in cold climates,  $\text{NO}_3^-$  concentrations in snow were found to not change with distance from tree trunks (Pomeroy et al., 1999), the samples from each location were assumed to represent throughfall and stemflow or bulk deposition with no redistribution from other areas. The SWE was measured and used as a surrogate for precipitation and samples were collected for nutrient analysis.

The samples were analyzed for  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , TDN and DOC concentrations. Dissolved organic N concentration was calculated by subtracting IN from TDN concentrations. When the IN concentration exceeded TDN (approximately 18% of the samples), DON was assumed to be zero. Ammonium,  $\text{NO}_3^-$  and DOC samples were filtered through a 0.45  $\mu\text{m}$  pore size Millipore filter within 48 h of collection and the  $\text{NH}_4^+$  and  $\text{NO}_3^-$  samples were preserved with sulfuric acid. TDN samples were filtered using a Whatman GF/F filter (mean pore size 0.7  $\mu\text{m}$ ). All samples were refrigerated at 4 °C until they could be analyzed (maximum storage time was three weeks). TDN samples were digested with potassium persulfate and reduced to nitrite in the presence of cadmium. Ammonium,  $\text{NO}_3^-$  and digested TDN samples were analyzed colorimetrically using a Lachat QuikChem 8500 FIA automated ion analyzer, whereas DOC samples were analyzed using a Shimadzu 5000A TOC Analyzer.

### 2.3. Data analysis

Daily nutrient fluxes ( $\mu\text{g m}^{-2} \text{day}^{-1}$ ) in bulk deposition and throughfall for the growing season and autumn were calculated by multiplying the amount of net precipitation (in mm) by the nutrient concentration (in  $\mu\text{g L}^{-1}$ ) of the sample and then dividing by the number of days since the sampler had last been emptied and cleaned. During the winter however, daily nutrient flux in bulk deposition and throughfall was calculated by multiplying the SWE by the nutrient concentration in each snow core and dividing by the age (in days) of the snowpack. This assumes no nutrient losses during the winter. Since stemflow volumes could not be accurately established, it was instead assumed that 6%, 1% and 0.4% of incoming precipitation was converted to stemflow by upland deciduous, upland conifer and wetland conifer stands, respectively, for the growing season and autumn (1 May to 31 October 2007). These values are within the range for the trees in these stand types reported in previous studies in other boreal and north temperate regions (Mahendrappa, 1990; Piirainen et al., 1998; Verry and Timmons, 1977). Stemflow was assumed to be negligible during the winter season. Therefore, daily nutrient flux in stemflow during the growing season and autumn was estimated by

multiplying the sample concentration by the estimate of stemflow volume and dividing by the number of days since the previous sample date.

For most dates with missing flux rates, linear interpolation was used to fill in gaps, although the daily flux rate for the last sampling date (11 October 2007) was assumed to remain constant until 31 October 2007. Annual flux rates were then calculated by summing the daily flux rates for the entire water year (1 November 2006 to 31 October 2007), while nutrient flow-weighted mean concentration was calculated by dividing the total nutrient flux by the total precipitation for three time periods: winter (1 November 2006 to 30 April 2007, estimated from the snowpack survey), growing season (1 May to 31 August 2007) and autumn (1 September to 31 October 2007). Throughout the manuscript, the term concentration refers to the flow-weighted mean concentration.

Daily nutrient flux rates and seasonal nutrient concentrations in bulk deposition and throughfall under the three canopy types (upland deciduous, upland conifer, wetland conifer and bulk deposition) were compared using a repeated measures analysis of variance (ANOVA). Annual flux rates and mean concentration for bulk deposition and the three stand types were compared using a single factor ANOVA. Multiple comparisons for significant ANOVA results were tested using the least squares difference and Scheffé's test. All statistical analyses were conducted using SPSS 16.0.

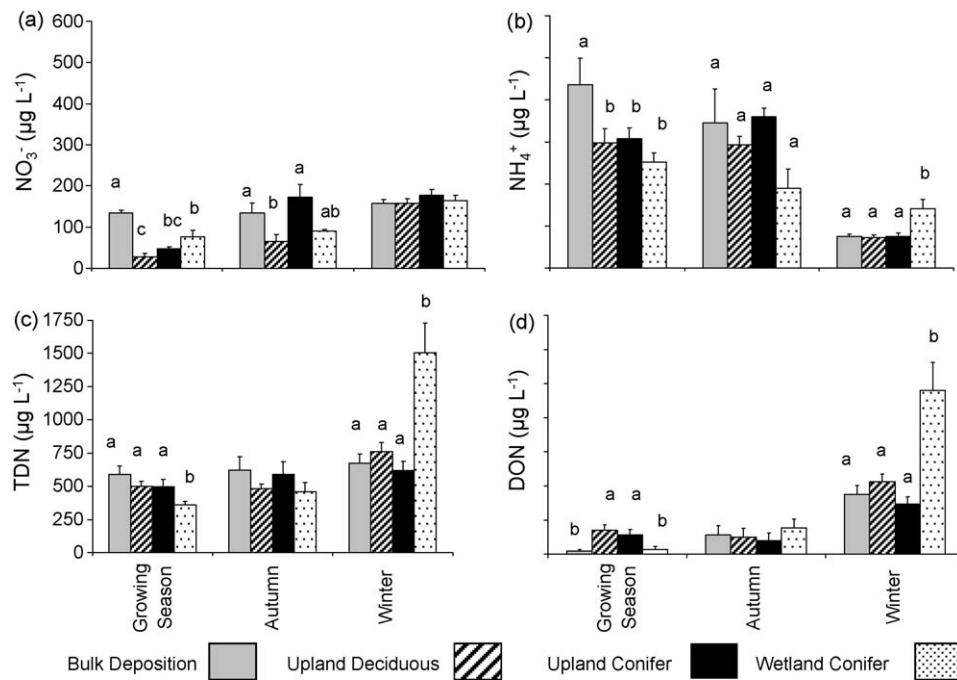
### 3. Results

The sampling year represented a typical year for the study area in terms of precipitation inputs as rain and snow. Total precipitation for the 1 November 2006 to 31 October 2007 water year was 547 mm at the Willow weather station. For comparison, total precipitation for the same time period was 582 mm at the Whitecourt weather station, approximately 50 km southeast of the study site, of which 25% fell as snow and 75% as rain (Environment Canada, 2008a). This is within 1% of the long-term (1971–2000) average annual precipitation for Whitecourt of 577 mm, with 24% falling as snow and 76% as rain (Environment Canada, 2008b).

Snow began to accumulate on 20 October 2006 and was completely melted by 15 April 2007 (Environment Canada, 2008a). Since the snowpack survey occurred 8 March 2007, approximately 13% of the snowfall was not included in the survey. Snowfall measurements at the Willow weather station indicate that approximately 5% of incoming snow in bulk deposition and upland stands was lost to sublimation or melting before the snowpack survey. Losses in the wetland conifer stands were higher, approximately 45% of incoming snow. Consequently the snowpack survey provided a good first approximation of winter throughfall.

The  $\text{NO}_3^-$  concentration was consistently higher in bulk deposition than in throughfall from all stand types during the growing season ( $P < 0.001$  for three stand types, Fig. 3a). The  $\text{NO}_3^-$  concentration in throughfall from all stand types was lowest during the growing season and highest in the winter snowpack, whereas the  $\text{NO}_3^-$  concentration in bulk deposition did not differ among seasons (Fig. 3a). During the growing season,  $\text{NH}_4^+$  concentration was also higher in bulk deposition than in throughfall from the three stand types ( $P = 0.01, 0.03$  and  $0.01$  for upland deciduous, upland conifer and wetland conifer, respectively, Fig. 3b). The  $\text{NH}_4^+$  concentration in the two upland stand types and the bulk deposition were lowest in the winter snowpack, while in the wetland stands the  $\text{NH}_4^+$  concentration was not detectably different by season (Fig. 3b). During the growing season, neither the  $\text{NO}_3^-$  nor the  $\text{NH}_4^+$  concentration differed among stand types.

There was no detectable difference in annual DON or TDN concentration between throughfall of the various stand types and bulk deposition ( $P = 0.26$  and  $0.34$ , respectively). However, during the growing season both upland stand types had throughfall DON



**Fig. 3.** Seasonal flow-weighted mean nutrient concentrations in bulk deposition and throughfall from three stand types for the Willow watershed. Note change in scale. Different letters above bars refer to differences ( $P < 0.05$ ) between stand types.

concentrations that were greater than the concentrations in throughfall of the wetland stands and in bulk deposition (Fig. 3c and d). The TDN concentration in the wetland stands were higher in the winter snowpack than in throughfall during the other seasons ( $P < 0.001$ , Fig. 3c), while TDN concentration in throughfall of the other stand types was not detectably different by season. In fact, during the winter, the DON and TDN concentrations for the snowpack in the wetland conifer stand types were approximately double the concentrations in the upland stand types and in bulk deposition (Fig. 3).

The DOC concentration in throughfall for all three stand types was higher than bulk deposition ( $P < 0.001$ , Fig. 4). Through most of the year, the DOC concentrations in throughfall were similar in the two conifer stand types (Fig. 4). For the winter period however, the DOC concentration was higher in the wetland than the upland conifer type (Fig. 4). Seasonal trends were also apparent in the two

upland stand types: the deciduous stands had the highest DOC concentration during the autumn, while the conifer stands had the lowest DOC concentration during the winter (Fig. 4). Both upland stand types had lower DOC concentrations during the winter compared to the other two seasons.

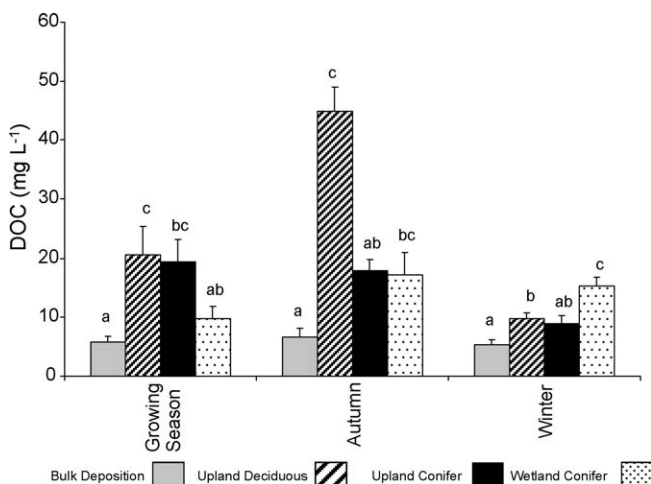
Estimates of daily flux rates for  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and TDN in throughfall and stemflow were less than flux rates in bulk deposition ( $P < 0.01$  for each, Fig. 5). Conversely, DOC daily flux rates were greater in throughfall and stemflow of the upland deciduous and conifer stands than in bulk deposition ( $P = 0.02$  and  $0.04$ , respectively), although there was no detectable difference between the throughfall of the wetland conifer stand and bulk deposition ( $P = 0.18$ , Fig. 5). There were no detectable differences between DON daily flux rates between any of the stand types and bulk deposition ( $P = 0.16$ ).

Annual  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and TDN flux rates to the forest floor of the three stand types were not detectably different, however the annual flux of all three N fractions below the canopy in all three stand types was consistently lower than the annual flux in bulk deposition ( $P < 0.001$  for each, Table 2). The estimates of annual DON flux rates in stemflow and throughfall of the three canopy types were similar to bulk deposition rates, although the DON flux was higher under deciduous stands than either the upland or wetland conifer stands ( $P = 0.04$ ).

In total, the annual IN flux below the canopy was approximately 50% of the IN flux in bulk deposition ( $P < 0.001$  for both  $\text{NO}_3^-$  and  $\text{NH}_4^+$ , Table 2). Annual TDN flux below the canopy was between 60% and 70% of the flux in bulk deposition whereas there was no detectable difference in annual DON flux between the throughfall of any stand type and bulk deposition (Table 2). Annual DOC flux was 2–3 times greater under all three forest canopies than in bulk deposition ( $P = 0.01$ , Table 3). Finally, the mean C:N mass ratio was 3–4 times higher in throughfall of all stand types than in bulk deposition.

#### 4. Discussion

The  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations in throughfall were approximately two-thirds of the concentrations in bulk deposition



**Fig. 4.** Seasonal flow-weighted dissolved organic carbon mean concentration in bulk deposition and throughfall from three stand types for the Willow watershed. Different letters above bars refer to differences ( $P < 0.05$ ) between stand types.

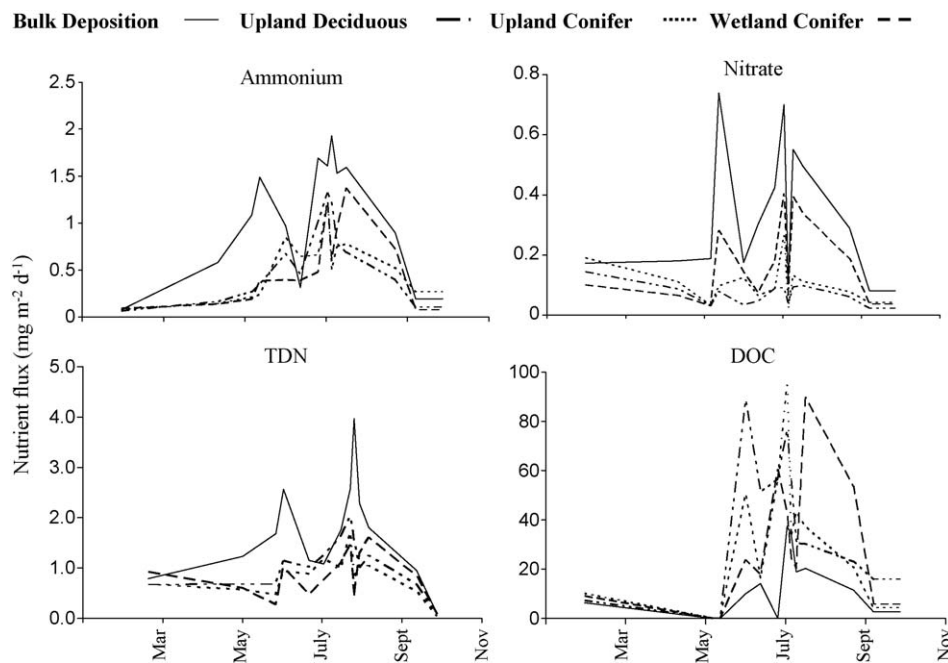


Fig. 5. Daily nutrient flux rates in bulk deposition and throughfall of three stand types for the Willow watershed. Note change in scale.

(Fig. 3), which suggests retention of IN by the forest canopy. The height of the trees and the multiple layers of foliage allow the forest to be much more efficient at intercepting dry deposition N than plastic funnels (Balestrini et al., 2007). As a result, the bulk deposition collectors likely underestimate the areal N deposition that occurs in the forest canopy. Thus, the amount of IN retained by the forest canopy may be greater than the data originally suggests.

Total dissolved N concentrations were similar between bulk deposition and throughfall. However since bulk deposition collectors are less efficient at intercepting dry deposited N than the forest canopy, TDN retention within the forest canopy is likely. In other words, the additional material retained in the forest canopy would be flushed through the canopy into the throughfall collectors during rain events, which would result in higher

concentrations in throughfall compared with bulk deposition for a given dry deposition rate. Since the bulk deposition and throughfall TDN concentrations were similar, it is likely that some N retention occurred within the forest canopy. However, since the relative proportions of wet and dry deposition of N to the forest canopy are unknown, the capacity of the canopy to retain TDN is uncertain.

The forest canopy retains N either through foliar uptake, absorption onto the leaf surface or assimilation by epiphytes and microorganisms within the canopy (Krupa, 2003; Lovett, 1994; Wilson, 1992). Wilson (1992) proposed that the mechanism for uptake is diffusion, however other research has noticed that N uptake is often correlated with a release of  $K^+$ ,  $Mg^{2+}$  and  $Ca^{2+}$ , as well as some weak organic acids (Draaijers et al., 1997; Lovett et al.,

Table 2

Mean  $\pm$  1 SE annual (November 2006 to October 2007) nitrogen flux above (bulk deposition) and below the forest canopy for three stand types in Willow watershed.

	Mean annual nitrogen flux ( $\text{mg m}^{-2} \text{ year}^{-1}$ )			
	Above canopy	Upland deciduous	Upland conifer	Wetland conifer
Nitrate				
Bulk deposition	80.3 $\pm$ 6.9			
Throughfall		32.8 $\pm$ 3.7	45.2 $\pm$ 1.7	40.6 $\pm$ 3.6
Stemflow		0.1 $\pm$ 0.0	0.1 $\pm$ 0.0	0.0 $\pm$ 0.0
Total below canopy		32.9 $\pm$ 3.7	45.3 $\pm$ 1.7	40.7 $\pm$ 3.6
Ammonium				
Bulk deposition	216.0 $\pm$ 10.1			
Throughfall		97.4 $\pm$ 6.0	94.9 $\pm$ 6.2	97.8 $\pm$ 9.0
Stemflow		6.0 $\pm$ 0.1	1.8 $\pm$ 0.2	0.6 $\pm$ 0.0
Total below canopy		103.4 $\pm$ 6.0	96.7 $\pm$ 6.4	98.4 $\pm$ 9.0
Dissolved organic nitrogen				
Bulk deposition	113.5 $\pm$ 10.7			
Throughfall		144.5 $\pm$ 11.7	96.9 $\pm$ 6.4	132.4 $\pm$ 13.8
Stemflow		14.2 $\pm$ 0.5	2.0 $\pm$ 0.3	0.4 $\pm$ 0.0
Total below canopy		158.7 $\pm$ 11.3	98.9 $\pm$ 6.3	132.8 $\pm$ 13.8
Total dissolved nitrogen				
Bulk deposition	409.8 $\pm$ 19.9			
Throughfall		274.7 $\pm$ 14.0	237.0 $\pm$ 7.7	270.8 $\pm$ 9.1
Stemflow		20.3 $\pm$ 0.5	4.0 $\pm$ 0.6	1.1 $\pm$ 0.0
Total below canopy		295.0 $\pm$ 13.8	240.9 $\pm$ 8.1	271.9 $\pm$ 9.0

**Table 3**Mean  $\pm$  1 SE annual dissolved organic carbon flux above (bulk deposition) and below the forest canopy for three stand types in Willow watershed.

	Mean annual flux ( $\text{g m}^{-2} \text{year}^{-1}$ )			
	Above canopy	Upland deciduous	Upland conifer	Wetland conifer
Bulk deposition	3.49 $\pm$ 0.64			
Throughfall		9.64 $\pm$ 1.58	7.80 $\pm$ 1.21	7.23 $\pm$ 1.38
Stemflow		0.78 $\pm$ 0.03	0.37 $\pm$ 0.05	0.14 $\pm$ 0.01
Total below canopy		10.42 $\pm$ 1.58	8.17 $\pm$ 1.18	7.38 $\pm$ 1.38

1985), suggesting active exchange of N for base cations. If N uptake in the forest canopy were strictly a function of diffusion of nutrients through the stomata, patterns of DON and IN retention or enrichment would be consistent. Instead, during the active growing season, the DON concentration below the canopy was higher than DON concentration in bulk deposition; opposite to the pattern observed for IN concentration (Fig. 3). The increased DON concentration in throughfall may be due to washing off of some dry deposited DON or leaching of weak organic acids (Draaijers et al., 1997). The inconsistent effect of the forest canopy on the different N fractions suggests that N uptake by the canopy is actively controlled, with preferential uptake of IN.

The increase in DOC concentration in throughfall (Fig. 4) suggests either leaching from the canopy or flushing of excess DOC that was deposited on the stems and foliage. Washing dry deposition off the foliage should not alter the C:N ratio, however the mean C:N ratio for throughfall from all stands was approximately 33, 4 times greater than the C:N ratio of bulk deposition. In remote areas, most of the atmospheric ON composition is organic nitrates, predominantly peroxyacetyl nitrate (PAN) (Neff et al., 2002). The C:N ratio of PAN is very low, leading to a low C:N ratio in bulk deposition. The increased C:N ratio in throughfall indicates a different source of ON than just atmospheric deposition, consistent with previous studies that measured leaching of organic acids from the foliage (Draaijers et al., 1997).

Similar to other studies in north temperate ecosystems the  $\text{NH}_4^+$  and  $\text{NO}_3^-$  concentration in throughfall was lower than the concentration in bulk deposition ( $P < 0.001$  for both N fractions, Table 2) (Duchesne and Houle, 2006; Friedland et al., 1991; Piirainen et al., 1998; Potter et al., 1991; Pryor and Barthelmie, 2005). Throughfall studies are not all consistent though, as some studies show enrichment of N in throughfall (e.g., Henderson et al., 1977; Mahendrappa and Ogden, 1973; Terauda and Nikodemus, 2007). The contrary results in throughfall N flux between various studies seem to be related to N deposition rates. Assimilation of IN by the canopy tends to occur in areas with low IN deposition rates (less than  $400 \text{ mg N m}^{-2} \text{year}^{-1}$ ), while N enrichment in throughfall occurs in areas with higher N deposition (Dise et al., 1998). The lower IN concentrations under the canopy therefore suggest that IN is still limiting primary productivity in this region. This is consistent with the low IN exports in surface waters from the Willow watershed (Pelster et al., 2008).

The concentrations of all N fractions and DOC were highly variable among sample dates (data not shown). Some of the variability in concentrations was due to the length of the sample period (increased dry deposition over time) and the volume and intensity of precipitation (dilution of the dry N deposition) (e.g., Chapin and Kedrowski, 1983; Morris et al., 2003; Shaw et al., 1989). Since dry deposition of N can be fairly high, ranging from 20% to 50% of bulk N inputs (Kelly and Meagher, 1986; Lovett and Lindberg, 1984; Sievering et al., 2000; Swank and Waide, 1987), much of the dry N deposition intercepted by the canopy may remain on the leaf surface until precipitation washes it through the canopy. As a result, the concentration of N in throughfall is typically highest at the beginning of storm events, with concentrations quickly decreasing as the event proceeds (Hill et al., 1999; Pryor and Barthelmie, 2005).

There were interesting seasonal patterns in the N concentration in the throughfall. The  $\text{NO}_3^-$  concentrations were higher in the winter snowpack than in throughfall during other seasons, while the  $\text{NH}_4^+$  concentrations were lower in the snowpack. There were no detectable seasonal changes in DON or TDN concentration. These findings were contrary to Morris et al. (2003), who found that DON concentrations of throughfall in northwestern Ontario black spruce stands approached zero during the winter. This difference from Morris et al. (2003) in winter DON concentrations was likely related to different sampling methods. To estimate N loading to the soils and surface water during the spring snowmelt, this study measured N flux for the winter period by sampling snow cores near the end of the season, whereas Morris et al. (2003) sampled the snow numerous times throughout the winter. Unfortunately, the remoteness of the site and the depth and lack of consolidation within the snowpack made this method unfeasible. However, since the TDN concentrations in bulk deposition and throughfall under the upland canopies do not differ by season (Fig. 3), any additional sources of DON to the snowpack (e.g., decomposition of forest litter within the snowpack) were likely minimal.

In the wetland snowpack however, the TDN concentration was higher than the concentration during other seasons, even more than would be expected given sublimation losses, and may indicate an additional source of N in these wetlands. Greater sublimation losses suggest longer retention of snow within the canopy leading to greater snow loads on the branches and longer contact time between the snow and foliage. Longer contact time may lead to additional N transfer from the foliage to the snow, while the increased snow loads may lead to increased loss of foliage and small shoots, which then end up in the snowpack. In addition, since DON concentration was estimated by calculating the difference between TDN and IN, the measurement error was potentially compounded.

The decreased  $\text{NH}_4^+$  and concurrent increased DON concentration suggests conversion of  $\text{NH}_4^+$  to DON within the snowpack. Although immobilization rates and temperature tend to be positively correlated there are some microorganisms that are adapted to snow-covered conditions and can immobilize IN throughout the winter period (Schmidt and Lipson, 2004). Therefore, these snow adapted microorganisms were likely responsible for the production of DON in the snowpack.

The TDN,  $\text{NH}_4^+$  and DON concentrations of the snowpack in wetland conifer stands were much higher (approximately double) than the TDN,  $\text{NH}_4^+$  and DON concentration of the upland stands and bulk deposition. Examination of the snowpack in the various stand types demonstrated that for the March snow survey, the SWE in the wetland stands was about 55% of the SWE in the other two stand types and open areas. The lower SWE and increased nutrient concentrations were probably related to higher interception and sublimation rates during winter for black spruce stands compared with either aspen or lodgepole pine stands (Pomeroy et al., 1998). Similar nutrient flux rates between stand types, combined with increased sublimation were likely responsible for the higher concentration.

The seasonal patterns for DOC concentration in throughfall were expected since other studies in boreal forests have detected similar



patterns (Morris et al., 2003; Verry and Timmons, 1977). Decreased concentrations during winter, even for the conifer stands, suggest that an active canopy allows more DOC through than a dormant one. This indicates additional sources of DOC in throughfall besides dry deposition; likely the leaching of weak organic acids, which is known to occur in the forest canopy (Draaijers et al., 1997). Increased DOC flux during the active growing season was consistent with other studies that found that the effect of the canopy on nutrient concentration was greatest during the active growing season (Henderson et al., 1977; Potter et al., 1991).

Similar to the one year of data collected previously by Shaw et al. (1989), most of the annual  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and TDN flux occurred during the growing season (Fig. 5). Since N flux in bulk deposition within the region was positively correlated with the amount of precipitation (Shaw et al., 1989), the large summer storms typical of the Boreal Plain result in greater N fluxes than during the rest of the year when precipitation is more limited.

Annual  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and TDN flux rates to the soil under the three stand types (i.e., the estimated sum of N in stemflow and throughfall) were not detectably different, however the total annual flux below any canopy type was consistently lower than the total annual flux in bulk deposition ( $P < 0.001$  for  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and TDN flux, Table 2). The lack of a difference by stand type is uncommon, since many other studies in temperate forests typically find differences by stand type (Henderson et al., 1977; Michalzik et al., 2001; Verry and Timmons, 1977). There was a fairly high degree of variation within each treatment type that may prevent detection of differences between the soil/stand types. Also, a meta-analysis of throughfall studies indicates that differences in throughfall IN flux between conifer and deciduous stands were much smaller where the annual N flux in bulk deposition is less than  $1000 \text{ mg N m}^{-2}$  (De Schrijver et al., 2007). Since the annual IN flux in bulk deposition for 2007 was only  $296 \text{ mg N m}^{-2}$ , the throughfall N flux in the different stands were likely to be much more similar than many of the previous studies that were conducted in areas of greater N deposition.

Annual bulk N deposition rates measured in Canadian boreal forests range from a low of  $230 \text{ mg N m}^{-2} \text{ year}^{-1}$  in northwestern Ontario to approximately  $1130 \text{ mg N m}^{-2} \text{ year}^{-1}$  in a study in central Ontario (Table 4). There appears to be a gradient from west to east, as higher flux rates were generally recorded in eastern Canada, although certain, more remote areas, such as northwestern Ontario still have low N loading rates (e.g., Morris et al., 2003). This gradient in atmospheric N deposition rates across Canada may be due to differences in climate and proximity to agricultural sources of  $\text{NH}_4^+$  and large sources of industrial and vehicle emissions of  $\text{NO}_x$ . For example, Kochy and Wilson (2001) found that total N deposition rates ranged from 680 to  $2210 \text{ mg N m}^{-2} \text{ year}^{-1}$  for various national parks in western Canada, with the greatest N deposition occurring within an aspen parkland forest surrounded by agricultural land and immediately downwind of a large urban and industrial center. In comparison, N deposition rates in the eastern U.S. are approximately  $1200$ – $1600 \text{ mg N m}^{-2} \text{ year}^{-1}$  (Table 4), while rates in parts of Europe can be as high as  $6400 \text{ mg N m}^{-2} \text{ year}^{-1}$  (Gundersen, 1995).

The N in bulk deposition was dominated by IN fractions, which comprised approximately 72%, while only 28% of the N in bulk deposition was DON. This composition was similar to the 30% suggested in a meta-analysis of North American and European studies (Neff et al., 2002). Organic N meanwhile was a major component of throughfall, comprising between 41% and 54% of N flux in all three stand, similar to what was found in North Carolina, where approximately 50% of N in throughfall consisted of DON (Quals et al., 1991).

The higher annual DOC flux in throughfall and stemflow were similar to a study in Minnesota where DOC flux was 6–12 times

**Table 4**

Annual nitrogen inputs ( $\text{mg m}^{-2} \text{ year}^{-1}$ ) in bulk deposition for regions across North America.

Location	Annual input	Source
New York (Whiteface Mountains)	1600	Friedland et al. (1991)
Indiana	1600	Pryor and Barthelmie (2005)
New York (Catskill Mountains)	1360	Lawrence et al. (2000)
Eastern Tennessee (Walker Branch)	1240	Kelly and Meagher (1986)
Central Ontario	1130	Molot and Dillon (2003)
Eastern Ontario	990	Devito et al. (1989)
North Carolina (Coweeta)	920	Swank and Waide (1987)
North Carolina (Coastal Plain)	580	Brinson et al. (1980)
South Carolina (Coastal Plain)	510	Richter et al. (1983)
Central Alberta	470–540	Alberta Environment (2008)
Central Alberta	424	Shaw et al. (1989)
Alberta (Boreal Plain)	410	This study
Northern Quebec	310	Duchesne and Houle (2006)
Central Saskatchewan	270	Huang and Schoenau (1997)
Northwestern Ontario	230	Morris et al. (2003)
Oregon (Western Cascades)	220	Stednick (2008)

greater under a canopy than in the open (Kolka et al., 1999). Unlike the Minnesota study however, the DOC flux in this study increased by only a factor of 2–3 (Table 3). This may be related to the low amount of precipitation at the study site during autumn (25.4 mm) when DOC concentrations were highest, compared with 260 mm of rain that fell during autumn during the Minnesota study (USDANRS, 2009). The annual DOC flux rates in bulk deposition (Table 3) were at the low end of the range ( $4$ – $16 \text{ g m}^{-2} \text{ year}^{-1}$ ) for 42 temperate sites across North America and Northern Europe (Kolka et al., 1999; Michalzik et al., 2001). The low annual DOC flux rates were likely related to the sub-humid climate since annual DOC flux rates in bulk deposition were positively correlated with annual precipitation (Michalzik et al., 2001).

## 5. Conclusions

Bulk deposition, throughfall and stemflow are important sources of labile N for forest vegetation; however no previous research has been conducted on the Boreal Plain to examine how a forest canopy alters the chemistry and flux of N to the soils. For an average precipitation year, the IN concentration in throughfall was lower than the concentration in bulk deposition. This decrease in IN concentration in throughfall compared with bulk deposition is consistent with a region where IN is limiting. However there were no detectable differences between the annual IN, TDN or DON flux rates of throughfall and stemflow in different soil/stand types. Instead, all canopies were equally able to remove N from bulk deposition, resulting in less N flux in throughfall and stemflow compared with bulk deposition. The inability to detect differences between the different stand types may be related to low N deposition rates in the region. The flux of DOC however, increased under the three forest canopy types, due to either leaching of organic compounds from the canopy or flushing of dry deposited DOC from the canopy.

The N inputs in bulk deposition were primarily IN, although DON comprised approximately 28% of the TDN. Dissolved ON however, composed a larger proportion (48%) of the throughfall flux. Seasonally, the canopy was much more active in taking up IN during the growing season and these effects were dramatically lower during the winter. In general, the annual N and DOC flux rates were low relative to other studies across North America and



Europe but similar to previous studies in the region (Shaw et al., 1989), suggesting that the impact of industrial and agricultural development, both known sources of atmospheric N, remains limited in this portion of the Canadian Boreal Plain.

Since annual N deposition in bulk deposition varies with precipitation (Shaw et al., 1989), this estimate of  $410 \text{ mg N m}^{-2} \text{ year}^{-1}$  is likely valid for years that also have precipitation rates similar to the annual average. Since more than 60% of the years on record have annual precipitation more than 10% away from the mean, it is likely that the bulk deposition and throughfall estimates are appropriate for approximately 40% of years. Therefore, although this study provides an estimate of bulk deposition and throughfall flux for an average year, many more years of data collection are required to understand the range of variation for N flux in bulk deposition and throughfall within this region.

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